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Computed contributions to odd nitrogen concentrations in the Earth's polar middle atmosphere by energetic charged particles

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Abstract

A two-dimensional photochemical transport model which has inputs that characterize the odd nitrogen production associated with galactic cosmic rays, solar particle events (SPEs), and lower thermospheric contributions (auroral electrons and solar EUV and soft X-rays) is used to compute odd nitrogen concentrations in the polar middle atmosphere from 1 January 1970 to 31 December 1994. We are able to separate out of the total odd nitrogen budget the contributions of the energetic charged particles according to type. The SPE contributions to annual average odd nitrogen concentrations in the polar stratosphere (latitudes > 50°) are computed to be significant (>10%) only for the larger events of August 1972 and October 1989. The SPE contributions to odd nitrogen concentrations in the polar middle atmosphere are found to be asymmetric with respect to hemispheres. The computed SPE contributions to odd nitrogen concentrations at 30 km are significant more often over the South Pole than the North Pole. The thermospheric contributions to odd nitrogen concentrations in the polar middle atmosphere are asymmetric with respect to hemispheres. A stronger thermospheric influence in the stratosphere is computed over the South Pole than the North Pole. An attempt has been made to compare the modeled odd nitrogen of the polar middle atmosphere to an ultra-high resolution polar ice cap nitrate sequence to examine the hypothesis that the nitrate sequences exhibit a signal associated with energetic particles. Variations of odd nitrogen production and modeled concentrations associated with energetic particles themselves cannot explain all of the huge variations observed in the fine structure present in nitrate data from the polar ice cap nitrates, but may be able to explain parts of some of them.

1. Introduction

The evaluation of the sources and concentrations of odd nitrogen chemical species (NO_y = N, NO, NO_2, NO_3, $2N_2O_5$, $BrONO_2$, $ClONO_2$, HO_2NO_2, and

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HNO_3) in the Earth's middle atmosphere is important for climate studies such as stratospheric ozone balance. The largest global source of odd nitrogen species in the stratosphere is the oxidation of nitrous oxide (Bates and Hays, 1967; Vitt and Jackman, 1996). Nitrous oxide (N_2O) is produced in the biosphere on the Earth's surface and is transported into the stratosphere where it reacts with $O(^1D)$, an electronically excited oxygen atom.

$$NO + O(^1D) \rightarrow NO + NO \tag{1}$$

$O(^1D)$ is produced in the stratosphere by the photodissociation of ozone.

$$O_3 + h\nu \rightarrow O(D) + O_2 \tag{2}$$

Odd nitrogen is also formed in the polar regions by incident solar and geomagnetic energetic charged particles, which deposit their energy in the Earth's middle and upper atmosphere. These energetic particles ionize and dissociate ambient O_2 and N_2 and initiate the formation of odd nitrogen. Since these particles are charged, they follow the Earth's magnetic field lines into the polar atmosphere. Changes in solar and geomagnetic activity can dramatically change the flux of incident energetic particles, e.g., solar particle events or auroral events, which lead to large variations of the formation of odd nitrogen in the polar middle and upper atmosphere (Vitt and Jackman, 1996). The variations of galactic cosmic ray particles related to solar cycle variations also modulate the formation of odd nitrogen in the middle and upper atmosphere (Vitt and Jackman, 1996).

It has been suggested that evidence of energetic particle events and solar cycle variations may be present in the polar ice sheets (e.g., Zeller and Dreschhoff, 1995). The history of the Earth's polar atmospheric chemistry is recorded by the chemical constituents which are present in stratified layers in the polar ice cap. The precipitating snow retains a signature of atmospheric chemical and physical conditions at the time the snow fell. It is also assumed that in ice sheets at latitudes above the melt line, the snow is retained in the ice sheet until glacier flow mechanisms transport the snow away or major climate changes occur that melt the snow. Hence, impurities stratified in the polar ice sheets such as nitrate ions (NO_3^-) provide a unique and very long-term record, of the order of hundreds of thousands of years, of environmental and climate change much like bottom lying sediments in oceans and lakes (Dansgaard et al., 1975; Oeschger, 1985; Dansgaard and Oeschger, 1989; Lorius et al., 1989; Mayewski et al., 1993, 1994). During a polar night, odd nitrogen is long-lived and may be transported down from the lower thermosphere and mesosphere into the stratosphere. Nitric acid (HNO_3), the terminal odd nitrogen species, is sequestered by polar stratospheric clouds which are formed in the low stratospheric temperatures of polar night (Tabazadeh et al., 1994; Song, 1994). These particulates which contain HNO_3 may then settle out of the stratosphere and become incorporated into the polar ice sheets possibly giving rise to nitrate ion anomalies observed in nitrate sequences extracted from the polar ice sheets.

Nitrates have been measured in polar snow and ice extracted from both the Greenland and Antarctica ice sheets (Wilson and House, 1965a, 1965b; Claridge and Campbell, 1968; Parker et al., 1977, 1978; Zeller and Parker, 1979; Laird, 1986; Dreschhoff and Zeller, 1990, 1994; Zeller and Dreschhoff, 1995). Ultra-high resolution ($> \sim 30$ samples/year) analysis of the nitrates in the Greenland ice record of more than 400 years shows a very sporadic history of nitrate anomalies (two standard deviations above the mean) superimposed on a background including seasonal variations (see Fig. 1).

In general, nitrate sequences observed in polar ice exhibit a seasonal variation in the nitrate concentration with the minimum occurring during the winter months and peaks occurring during the summer months. The summer peaks in the nitrate sequences have been pointed out by Finkel et al. (1986), Neubauer and Heumann (1988), Steffensen (1988), Davidson (1989), Whitlow et al. (1992), Mulvaney and Wolff (1993), Yang et al. (1995), and many others. It has been suggested that the conditions of full summer sunlight and the period of highest temperature lead to an increase in sublimation resulting in an increase of nitrate concentration in the snow surface (Laird, 1986; Laird et al., 1987; Dreschhoff and Zeller, 1994).

Based on correlation of nitrate concentration peaks and solar activity measures, it has been suggested that observed nitrate sequences in the polar snow and ice may be controlled, at least to some degree, by solar activity (Zeller and Parker, 1981; Laird et al., 1982; Laird, 1983; Dreschhoff et al., 1983, 1993; Dreschhoff and Zeller, 1990, 1994; and Zeller and Dreschhoff, 1995). It has been suggested that nitrate enhancements caused by large sequence solar particle events (SPEs) originating near the central meridian of the sun are distinguishable from meteorologically derived nitrate peaks (Shea et al., 1993). It has been postulated by Dreschhoff, Zeller, Laird, and others that the nitrate signal retained in the stratigraphy of the ice sheets is associated with atmospheric ionization and dissociation caused by incident energetic particles driven by solar and geomagnetic events. Zeller and Parker (1981) also suggest that there may be solar cycle variations in South Pole nitrate sequences. Zeller and Parker present raw nitrate data collected from the South Pole and Vostok, located ~1300 km apart. Both South

Pole and Vostok data sets exhibit a drop in nitrate content at a depth which Zeller and Parker have associated with the Maunder minimum. With the spikes of these data sets removed, the background data sets yielded a linear coefficient of 0.69. Zeller and Parker, therefore, conclude that variations in these nitrate sequences probably have a common origin despite their 1300 km separation. Frequency analysis of both data sets show strong 65- and 22-year periodicities. 22- and 11-year periodicities were clearly present when the well-dated portion of the sequences were examined (Dreschhoff et al., 1983).

Early model studies of the response of the NO_y budget above the tropopause to solar variations have been performed (Garcia et al., 1984; Jackman et al., 1990; Legrand et al., 1989). Using a two-dimensional photochemical transport model Jackman et al. (1990) were unable to reproduce the large variabilities in the nitrate sequence reported by Zeller et al. (1986). Jackman et al. (1990) report a maximum NO_y enhancement of about 10% above the modeled background at 75°S associated with the August 1972 SPE. Assuming that the entire amount was transported to the surface within one month, Jackman et al. estimated the corresponding nitrate deposition flux to be 1.6 mg m^{-2} NO_3^- $month^{-1}$, which is in reasonable agreement with Zeller et al. (1986). However, the nitrate maximum can only be 10% above the background and the enhancements observed by Zeller et al. are ~400% above the background for data with a time resolution of about one month. Jackman et al. conclude, "that the correlation of the spike with the August 1972 SPE is fortuitous and that the spike has a different origin." Other models (Legrand et al., 1989) report smaller percentage variations in the NO_y budget of the lower stratosphere in response to the particle precipitation events and other solar variations than the percentage variations in ice and snow (Zeller et al., 1986). It is recognized that the annual, model-derived NO_y values are based on stratospheric gas-phased reactions whereas the stratospheric nitrate (NO_3^-) found in polar ice as part of the total nitrate budget is thought to result from heterogeneous reactions in the winter polar atmosphere. The resulting nitrate signal reported by Zeller and Dreschhoff (1995), is found as part of the fine structure of the high-resolution data from Antarctica and Greenland

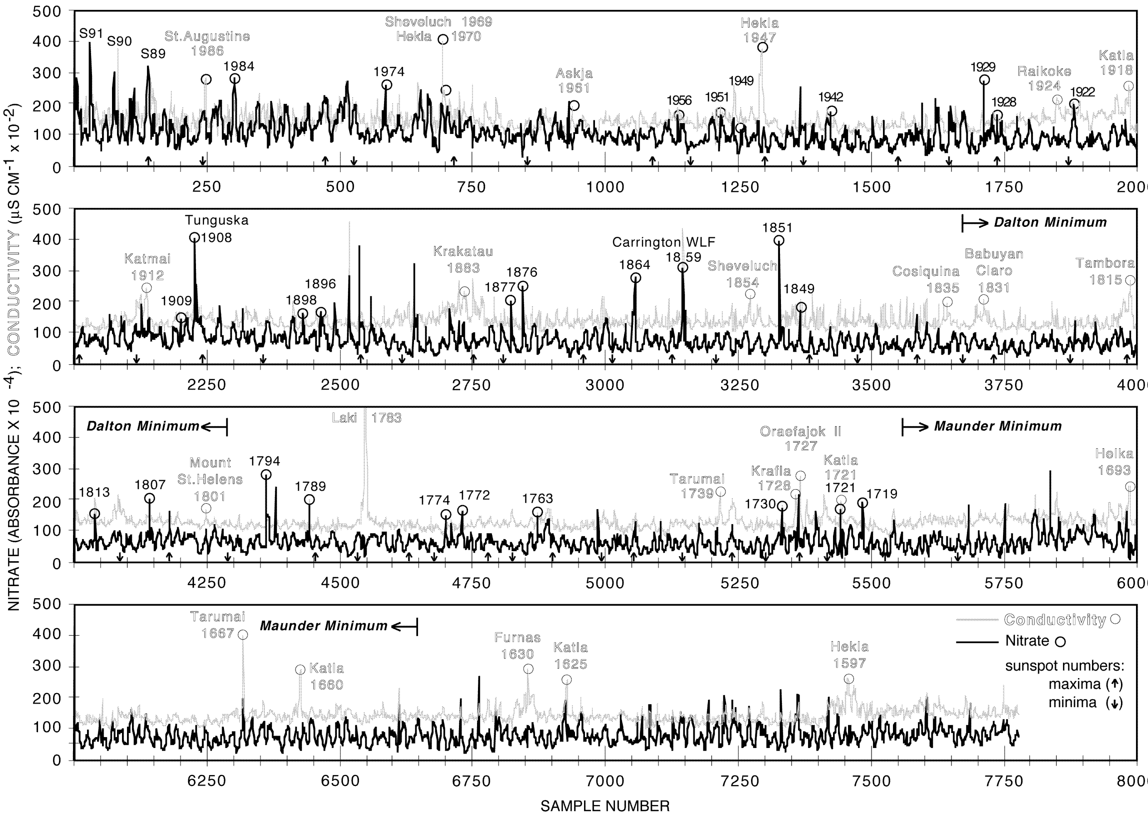

Fig. 1. Taken from Fig. 1 of Zeller and Dreschhoff (1995). Nitrate concentrations extracted from the central Greenland ice sheet. Also shown is the measured conductivity. Anomalies in the conductivity are used as a dating mechanism.

(Dreschhoff et al., 1997). This signal cannot be resolved in ice records of lower resolution data ($\geq$1 year). Multiple ice cores from both polar regions will provide a signal of highest quality for the evaluation of contributions from solar flares. Whereas this work is currently in progress (Dreschhoff et al., 1997),the modeled annual average odd nitrogen in this study is confined to the comparison with nitrate in Greenland ice which has been converted to annual values.

Legrand and Delmas (1986), Legrand and Kirchner (1990), and Wolff (1995) proposed a weak contribution to the polar ice cap nitrate sequences from middle atmospheric sources in comparison to tropospheric sources such as lightning at low and middle latitudes. Lightning is the main source of NO_x (which is composed of N, NO, and NO_2) that originates within the troposphere (Wolff, 1995). Because of the rising motion of air in the tropics, tropospheric NO_x produced in the tropics by lightning may be transported to the stratosphere (Ko et al., 1986; Wolff, 1995). Legrand and Kirchner (1990) have suggested that NO_x produced in the middle and low latitudes by tropospheric lightning may be transported poleward through the stratosphere and may account for a third to one half of the total NO_3^- content of the Antarctic snow and pre-industrial Greenland snow. The model results published by Ko et al. (1986) have shown that if only 5% of the tropospheric NO_y produced by lightning is transported to the lower stratosphere, this would amount to about one third of the global production produced by the oxidation of nitrous oxide in the stratosphere. All these processes may contribute to the background found in snow in both polar regions.

As briefly outlined above, the source of the background nitrate observed in the polar ice sheets is still unknown. Zeller, Dreschhoff and Laird have argued that there is a solar signal superimposed on the tropospheric nitrate background based on correlations of SPE sources and nitrate concentrations as well as approximate 22- and 11-year solar cycle periodicities. These are the results of the only nitrate sequences that are totally sequential without any gaps in the data and at ultra-high resolution. What has yet to be explained is the absolute quantitative relationship of variations in the sources of atmospheric ionization due to solar activity and the variations in deposited nitrate in polar snow. The investigations described above, when using relatively low resolution nitrate data, have not found a simple direct relationship between polar atmospheric odd nitrogen concentrations and ice nitrate concentrations. An additional complicating factor is that the variations in the ionization sources due to solar activity only appear to affect the additional atmospheric odd nitrogen concentrations by appreciable amounts on rare occasions. In addition, the transport processes of odd nitrogen from the middle atmosphere and the deposition processes into the polar snow are not well understood. We use a two-dimensional photochemical transport model with the goal of determining the relative contributions of galactic cosmic rays (GCRs), SPEs, and auroral electrons to the odd nitrogen budget of the polar middle atmosphere over a 25-year period. An attempt will be made to compare the model calculations to the upper portions of nitrate sequences extracted from the Greenland ice cap.

2. Modeling

2.1. Basic 2D model

We used the NASA/Goddard Space Flight Center two-dimensional zonally averaged photochemical transport model (GSFC 2D model) which is described by Vitt et al. (2000).

2.2. Characterizing the energetic particles

In addition to the ambient nitrous oxide oxidation source of odd nitrogen in the middle atmosphere Eq. (1), energetic charged particle sources are characterized and added to the basic GSFC 2D model. The various types of energetic charged particles considered in this analysis are galactic cosmic ray particles, solar particles, auroral electrons, photoelectrons and soft X-rays in the lower thermosphere. As described in previous papers (Vitt and Jackman, 1996; Vitt, 1997; Vitt et al., 2000), the power inputs into the model associated with these energetic particle events are varied based on time series of observations. As in Vitt and Jackman (1996), we assume that 1.25 molecules of NO_y are produced per ionization associated with these energetic particles. Solar particle inputs into the model are the satellite observations of the solar proton and alpha particle fluxes in the near Earth space environment (IMP 6 and IMP 8). These incident energetic particles are input into an energy deposition calculation to evaluate ionization profiles (Vitt and Jackman, 1996; Vitt, 1997). The GCRs are quantified using ionization observations of Nicolet (1975). The GCRs are modulated by use of the observed sun spot numbers as a proxy (Vitt and Jackman, 1996). The auroral electron inputs are modulated by use of satellite observations of the auroral electron hemispheric power (and by the Kp index for the time periods when satellite observations are not available) (Vitt, 1997; Vitt et al., 2000). Ionization profiles associated with the auroral electrons are determined by the Fuller-Rowell and Evans (1987) statistical model. Production profiles of Barth (1992) are used to estimate the odd nitrogen production associated with solar EUV photons and soft X-rays in the lower thermosphere which are modulated using the solar $F_{10.7}$ index (Vitt, 1997; Vitt et al., 2000). In Fig. 2 typical globally and yearly averaged odd nitrogen production profiles associated with these sources show their altitude locations.

The production of NO_y associated with relativistic electrons is not considered in this model.

3. Simulations

The GSFC 2D model has two modes of operation, steady state and time dependent. Steady state simulations of the 2D model are computations with fixed boundary conditions. The constituent concentrations calculated by these simulations reach a repeating annual cycle. All steady state simulations in this study are for boundary conditions corresponding to 1970. Time dependent simulations are computations where the bottom boundary conditions are determined as functions of time. The time dependent simulations used in this study are for conditions corresponding to a 25-year period, from 1 January 1970 to 31 December 1995. The time dependent boundary conditions for the source gases in the model simulations were taken from Table 6-3 of WMO (1995).

We are able to compute the relative contribution each energetic particle source makes to the odd nitrogen budget by separately turning off the individual sources and comparing the results with those of a simulation where all sources of NO_y are turned on. It is assumed that there are no non-linear interactions between these source processes. Four time dependent simulations are used to compute the contributions: (a) with all sources on, (b) same as (a) except GCRs turned off, (c) same as (a) except SPEs turned off, and (d) same as (a) except lower thermospheric sources turned off. The lower thermospheric sources include auroral electrons, photoelectrons, and soft X-rays. Initial conditions for these four simulations were prepared by four different appropriate time independent simulations which have 1970 boundary conditions.

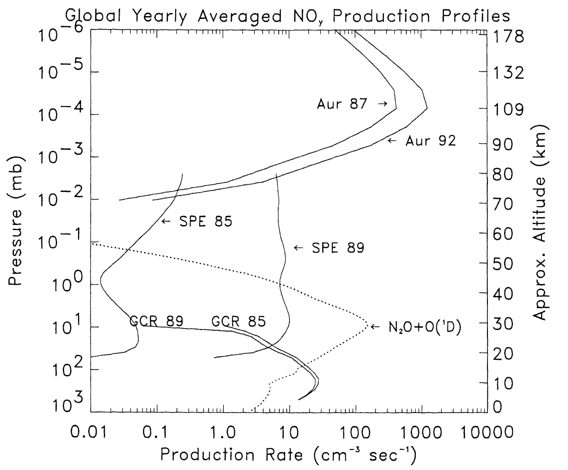

Fig. 2. Typical globally and yearly averaged odd nitrogen source profiles associated for years of both high and lower solar activity. Auroral electron source (Aur 87 and Aur 92) is located in the lower thermosphere; solar particle source (SPE 85 and SPE 89) in the mesosphere and upper stratosphere, which is highly variable; galactic cosmic ray source (GCR 85 and GCR 89) in the lower stratosphere and upper troposphere; while the ambient biogenic source ($N_2O + O(^1D)$) dominates the global stratosphere.

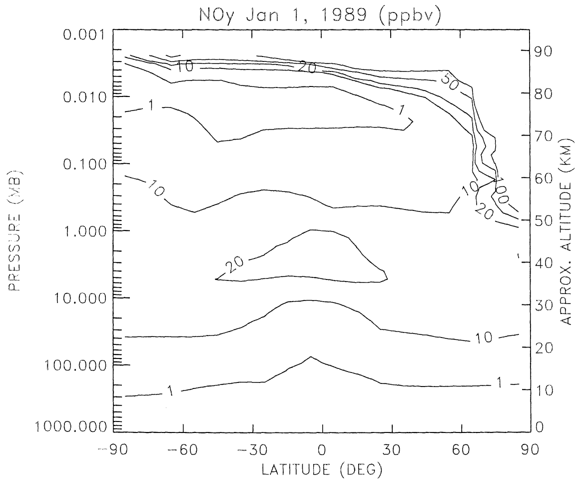

Fig. 3. Modeled mixing ratio of NO_y on 1 January 1989.

Figs. 3 and 4 show the NO_y mixing ratio fields at 1 January 1989 and 1 January 1990, respectively. Note the augmentation of stratospheric NO_y over both poles on 1 January, 1990, which is the result of the large

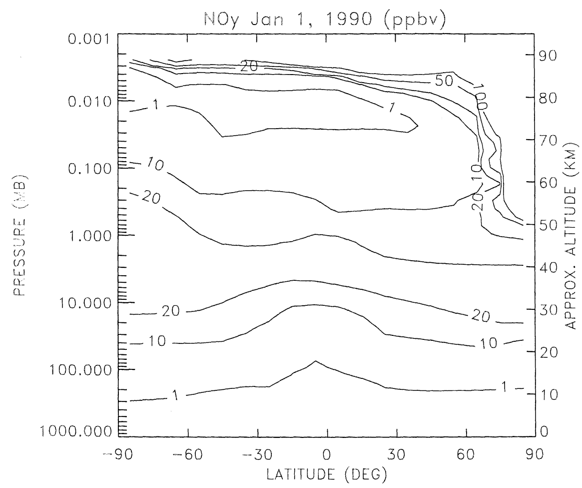

Fig. 4. Modeled mixing ratio of NO_y on 1 January 1990.

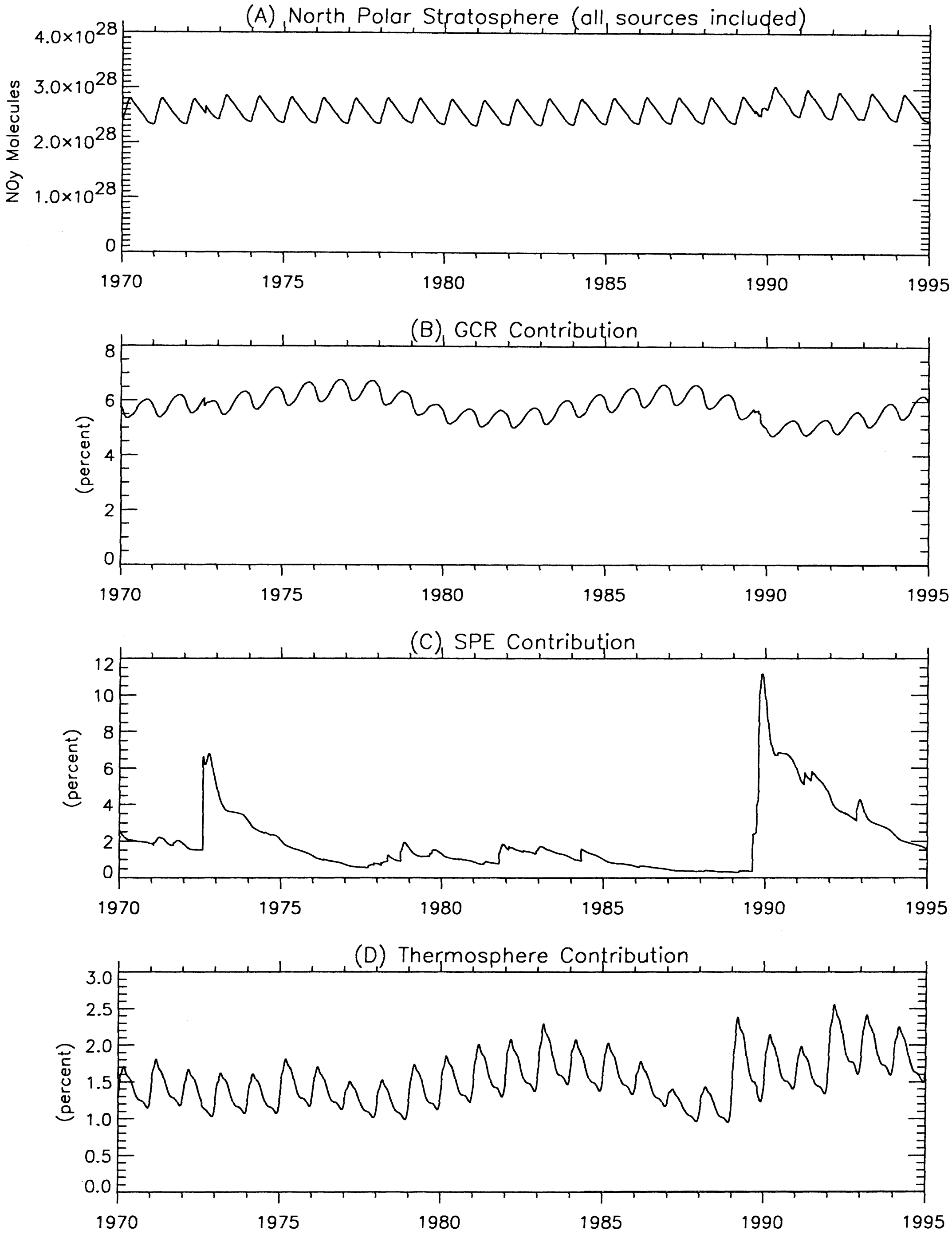

Fig. 5. The NO_y budget of the north polar stratosphere (50–90°N latitude, tropopause — 50 km). Panel (A) shows the total number of NO_y molecules. The contribution to this NO_y budget by GCRs is in (B), by SPEs is in (C), and by lower thermospheric processes is in (D).

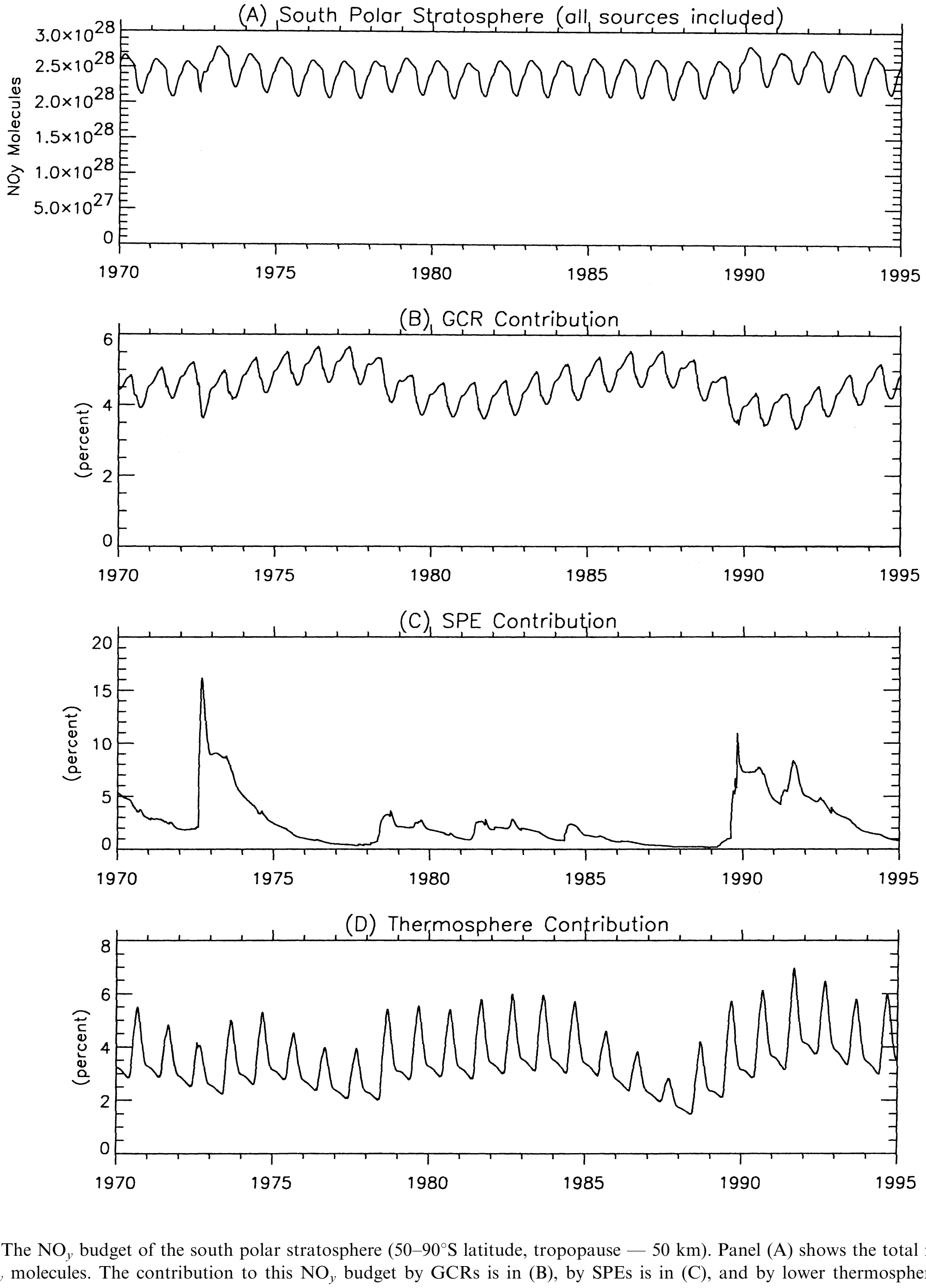

Fig. 6. The NO_y budget of the south polar stratosphere (50–90°S latitude, tropopause — 50 km). Panel (A) shows the total number of NO_y molecules. The contribution to this NO_y budget by GCRs is in (B), by SPEs is in (C), and by lower thermospheric processes is in (D).

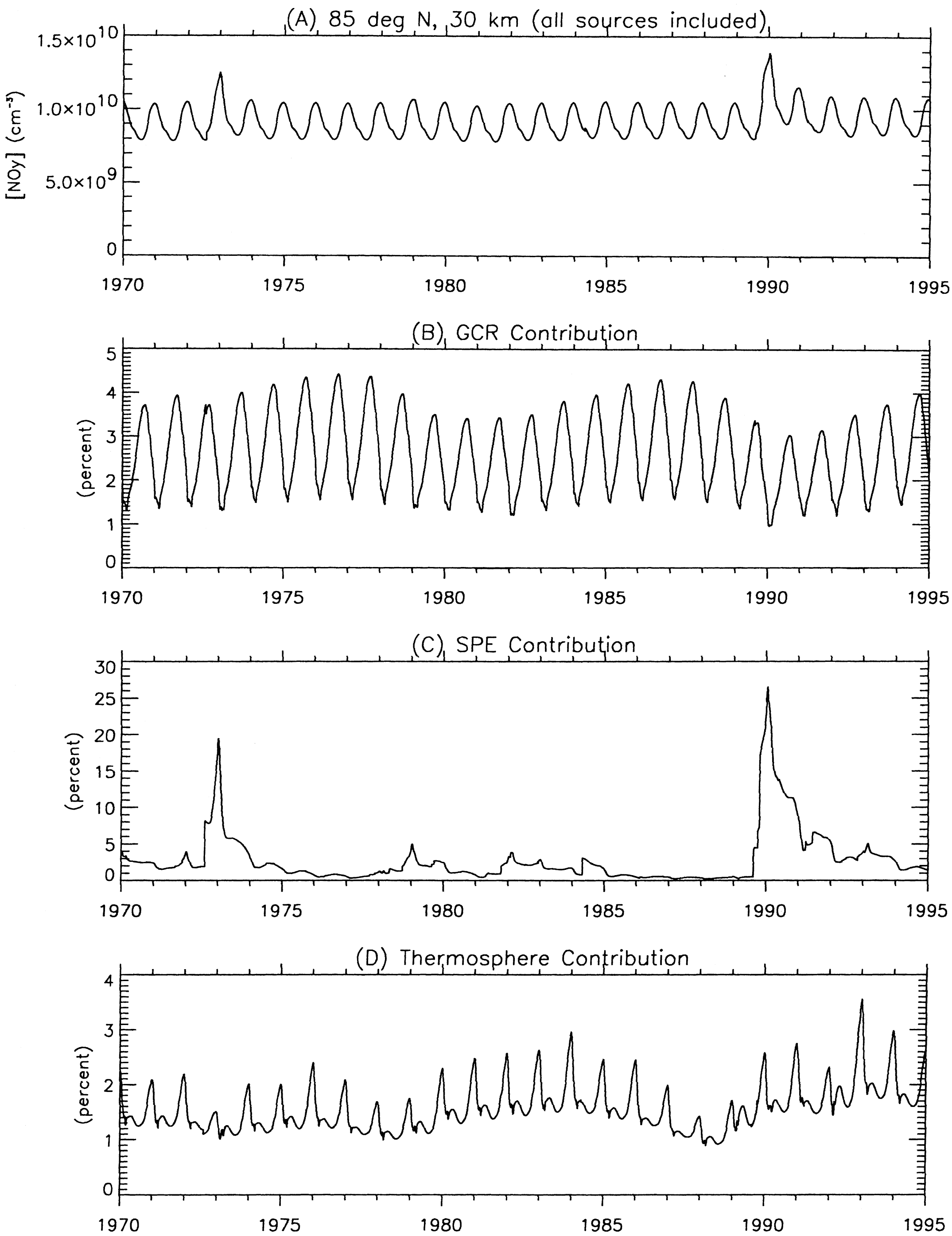

Fig. 7. The evolution of the NO_y concentration at a grid point located at 85°N and 30 km (A) from 1 January 1970 to 31 December 1994. The contribution to this NO_y budget by GCRs is in (B), by SPEs is in (C), and by lower thermospheric processes is in (D).

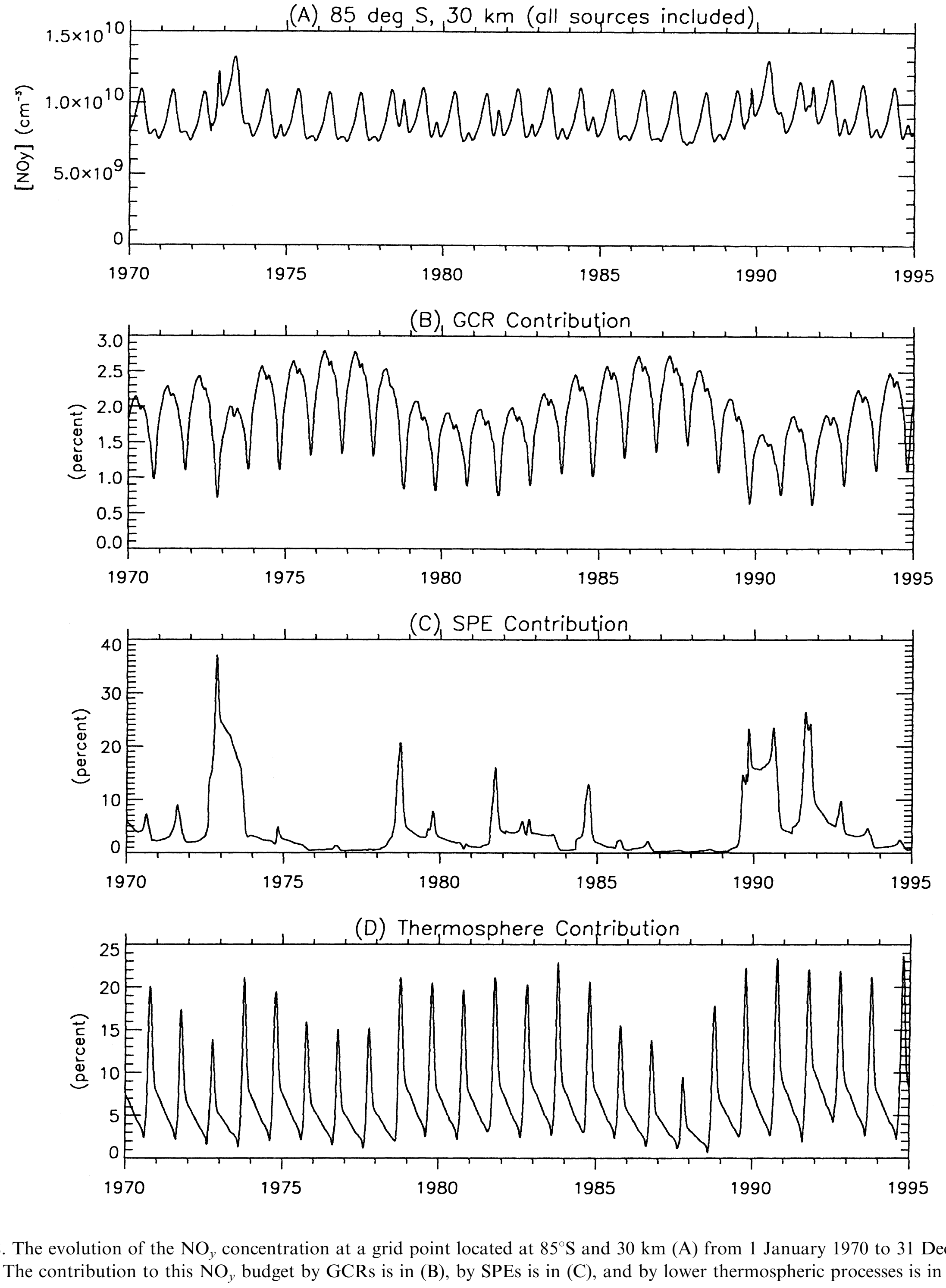

Fig. 8. The evolution of the NO_y concentration at a grid point located at 85°S and 30 km (A) from 1 January 1970 to 31 December 1994. The contribution to this NO_y budget by GCRs is in (B), by SPEs is in (C), and by lower thermospheric processes is in (D).

October 1989 SPE. Note also the tongue of NO_y that is transported down from the lower thermosphere during polar night.

3.1. Computed polar NO_y variations

Curve A of Figs. 5–8 shows the evolution of NO_y budget of the polar middle atmosphere from 1 January 1970 to 31 December 1994. These are the results of a time dependent simulation of the 2D model, which includes the production of odd nitrogen associated with GCRs, SPEs, and processes of the lower thermosphere. Figs. 5(A) and 6(A) are the total number of NO_y molecules in the North and South polar stratospheres, respectively, integrated from 50 to 90°N latitude and from the tropopause to 50 km. Fig. 7 shows the evolution of the NO_y concentration at 30 km, 85°N; Fig. 8 shows the same at 30 km, 85°S.

Curves B through D of Figs. 5–8 are the contributions by GCRs, SPEs, and lower thermospheric processes (auroral electrons and photoelectrons associated with the absorption of solar EUV and soft X-rays), respectively.

The 11-year solar cycle is evident in the GCR contribution curves. The GCRs are modulated by solar cycle activity. Also computed is an annual variation superimposed on the 11-year variation. The GCR contribution to the integrated NO_y budgets of the polar stratospheres (integrated from the tropopause to 50 km, and from 50 to 90° latitude) is of the order of 3–7% (see Curve B of Figs. 5 and 6).

The largest energetic particle contribution to the odd nitrogen budgets integrated over the polar stratospheres (50°–90° latitude, tropopause ~50 km) is by the SPEs during the August 72 and the October, 89 events (see Curve C of Figs. 5 and 6). This contribution to the total number of NO_y molecules in the South polar stratosphere reaches ~15% resulting from the August'72 event. Note, for the August'72 event we have chosen to use the larger hourly averaged integral fluxes of *The Geophysical Solar Publication* rather than the daily averaged integral fluxes of IMP 6 (see Jackman et al., 1990). The contribution is ~11% in the South Polar stratosphere from the October 89 event. The SPE contribution is asymmetric with respect to hemispheres because both transport and the photochemistry of NO_y depend on season, and the seasons of the two hemispheres are 6 months out of phase. Henceforth, the relative contributions depend on the season during which an event occurs. Also evident in the SPE contribution curves is the long-lived nature of the effect of the large events on the polar stratosphere in terms of standard photochemistry and gas phase only processes, of the order of a few years. This is because NO_y is long-lived in this region of the atmosphere.

An asymmetry is computed in the thermospheric contributions to the NO_y budgets in the polar regions. The thermospheric contributions to the integrated NO_y budget are stronger in the South Polar region than in the North Polar region. The thermospheric contribution to NO_y reaches ~2.5% in the North Polar stratosphere (see Fig 5(D)) and ~6% in the South Polar stratosphere (see Fig. 6(D)).

Fig. 7 shows NO_y and the relative contributions at 85°N, 30 km altitude. The GCR contribution reaches ~4.4% (see Panel B). The SPE contribution reaches ~27% (see Panel C). The effects of the SPEs are long-lived at this altitude as compared to the effects at 50 km, which are not shown here. Panel D shows that the thermospheric contribution reaches ~3.6%. Fig. 8 shows NO_y and the relative contributions at 85°S, 30 km altitude. The GCR contribution reaches ~2.8% (see Panel B). The SPE contribution reaches ~38% (see Panel C). Panel D shows that the thermospheric contribution reaches ~25%, which is much greater than the contribution computed at 85°N. Due to asymmetries in the transport parameters, a stronger down flux of thermospheric NO_x is computed in the southern hemisphere than in the northern hemisphere.

In summary, relative contributions to NO_y in the polar middle atmosphere by GCRs, SPEs, and thermospheric sources have been quantified. The following points have been shown in this study:

- The GCR contributions exhibit a solar cycle modulation.
- The SPE contributions to odd nitrogen concentrations are asymmetric with respect to hemispheres — the photochemistry and transport are seasonally dependent.
- The SPE contributions to odd nitrogen concentrations are significant more often at 30 km over the South Pole than over the North Pole.
- The thermospheric contributions to odd nitrogen concentrations in the polar stratosphere are asymmetric with respect to hemispheres — there is a stronger thermospheric influence in the southern hemisphere.

4. Comparison with the nitrate sequences

Given the sporadic nature of the major anomalies, as defined by Shea et al. (1993), in ultra-high resolution nitrate sequences, it might be possible that these anomalies are associated with the sporadic sources of odd nitrogen in the polar atmosphere. These sporadic sources are located above the tropopause. An attempt has been made to compare the observed polar ice nitrate sequence of Dreschhoff and Zeller (1994) and

Zeller and Dreschhoff (1995) extracted from Greenland ice cores with the modeled odd nitrogen concentrations in the stratosphere above the North Pole (Fig. 9).

The modeled NO_y concentrations at 85°N at an altitude of 30 km as a function of time are shown in Fig. 9(A). This is the result of the 25-year simulation which begins 1 January 1970, and runs through 31 December 1994, the same concentrations as shown in Fig. 7(A).

The background concentrations may need to be removed from the NO_y budget before a comparison can be made with the nitrate sequences. Fig. 9(B) shows the computed NO_y budget at 30 km at 85°N with the background removed. This is the contribution to the NO_y budgets made by the SPE and thermospheric sources — the difference between a simulation with all sources and a simulation without the sporadic energetic particle sources (SPE and thermospheric NO_y sources). However, the nitrate data in the upper portion of the sequence, as shown in Fig. 9(C), cannot serve direct correlation studies with modeled NO_y time series, because the data points are presented as sample numbers along the ice core and they represent a varying time scale due to varying snow accumulation and compaction. Therefore, the nitrate signal must be reduced to a similar time scale as that in the model. Fig. 10 shows annual nitrate based on the dating of the sequence by Gladysheva and Dreschhoff (1996). This data series displays a signal variation comparable to the general variation in the computed SPE contributions (Fig. 7C and 10B) as well as to the general, solar cycle dependent, number of relativistic SPEs (ground level events) (Shea and Smart, 1990). Furthermore, the nitrates as shown have been computed without removal of the background, which in this part of the sequence contains an upward trend from varying anthropogenic contributions. Fig. 10(A) also shows smoothing of the annual nitrate by a three-point moving average. Assuming that the smoothing may represent the general background as a first approximation, the variation above the background may be viewed as the superimposed SPE derived sig-

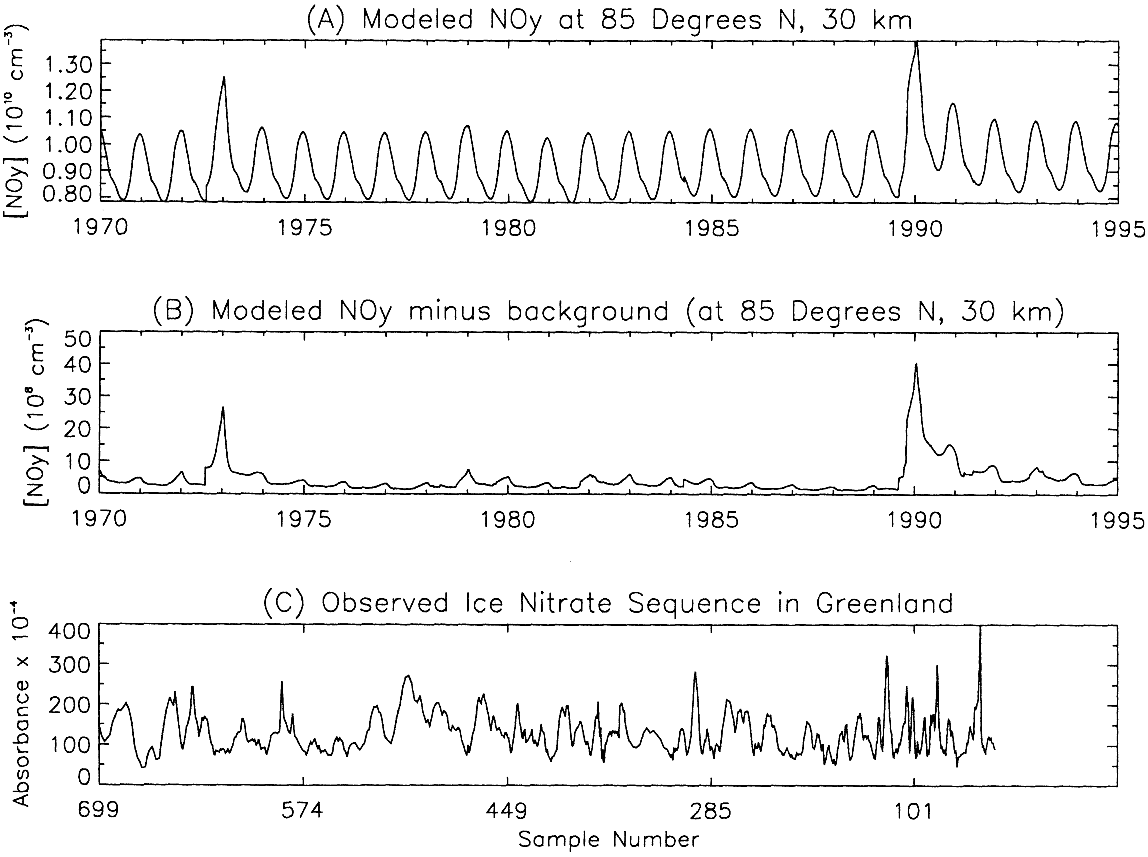

Fig. 9. Modeled concentrations of NO_y at 85°N at an altitude of 30 km versus time (A); same as (A) minus the modeled background NO_y (B); and the upper portion of the nitrate sequence extracted from the Greenland ice sheet Dreschhoff and Zeller, 1994; Zeller and Dreschhoff, 1995) which approximately corresponds to the 25-year simulation period (C).

nal. This signal falls into the range of about 27% for the year 1989, and about 19% for the year 1972. Further quantitative analysis of modeled SPE NO_y production and nitrate measurements in ice cores near both poles needs to be undertaken to prove or disprove the suggestive correlations discussed here.

4.1. Deposition of nitrates

Two mechanisms are possible for transporting NO_y from the polar stratosphere and depositing it into the polar ice caps as nitrate ions, NO_3^-. The mechanisms are associated with (1) conventional stratospheric–tropospheric exchange and (2) polar stratospheric clouds (PSCs). Through conventional stratospheric-tropospheric exchange, the NO_y will arrive along with other constituents. From the troposphere, the NO_y may then be either snowed out or directly deposited into the ice cap. NO_y can also be drawn out of the polar stratosphere by a process associated with the sequestering of nitric acid, HNO_3, by PSCs, which settle out of the stratosphere and become incorporated into the ice cap.

It is known that if ions are present in a saturated vapor, aerosol particles are formed at saturation ratios lower than those required for homogeneous nucleation (Adachi et al., 1992). This phenomenon is commonly referred to as ion-induced nucleation. It has been postulated that ionization events in the polar middle atmosphere induce nucleation of sulfuric acid (H_2SO_4) vapor forming condensation nuclei (CN) at altitudes of

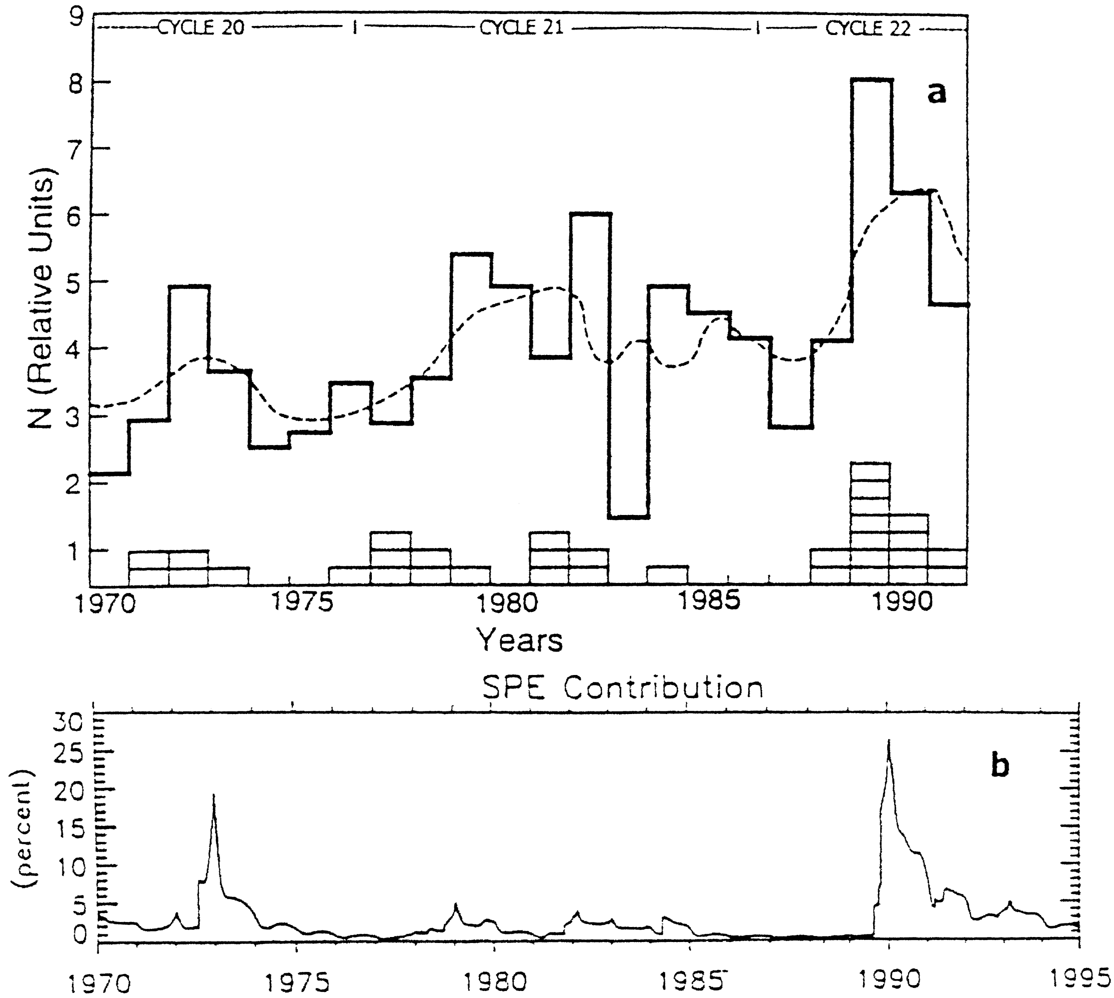

Fig. 10. (A) shows total yearly nitrate flux in relative units for the central Greenland ice sheet (72°N, 38°W) and the smoothed three-point averaging signal superimposed (adapted from Gladysheva and Dreschhoff (1996), in comparison with the number of relativistic solar proton events per solar cycle, each box representing one event Shea and Smart, 1990. These data are compared to the modeled SPE contributions (B) as shown in Fig. 7(C).

30–35 km (Arnold, 1982; Hofmann and Rosen, 1982, 1983a, 1983b). Some of these CN may further act as condensation centers for the formation of PSCs which uptake HNO_3 and settle out of the stratosphere in the polar regions. An enhancement of stratospheric aerosols after a solar particle event has been reported by Shumilov et al. (1996). Lidar measurements at Verhnetulomski observatory (68.6°N, 31.8°E) detected an increase of stratospheric aerosol concentration that seemed to be coincidentally occurring after an SPE. The temperatures at this location in the stratosphere were low enough to support the formation of PSCs; thus, it is unclear if there is a cause-effect relationship between the onset of a SPE and the production of aerosols. Clearly, more work needs to be carried out with other SPEs to determine if these events can have an influence on the formation of stratospheric aerosols.

5. Conclusions

The cause of the polar ice cap nitrate variations at the level of the nitrate fine structure cannot be explained by our middle atmospheric photochemical transport model. On the other hand, a preliminary comparison can be made if the nitrate data are transformed to the time scale of one year as employed by the model. This removes or filters the high frequency peaks which are a feature of the upper unconsolidated firn and reflect in part depositional variation such as monthly meteorological contributions (Dreschhoff and Zeller, 1990). As shown in Fig. 10(A), however,the total yearly nitrate signal can be used for a qualitative comparison with the modeled data in Fig. 10(B). The concept or working hypothesis of a one-to-one quantitative relationship between an SPE and nitrate concentration anomaly is being investigated further, because Shea et al. (1993) found apparent correlations that were unlikely to be due to coincidence. Since their data originates from greater depth in the core and more highly consolidated firn, they will provide a higher signal to noise ratio, i.e., meteorological noise is significantly reduced.

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